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CA 2415388 A1 2002/01/17

(21) **2 415 388**

(12) **DEMANDE DE BREVET CANADIEN
CANADIAN PATENT APPLICATION**

(13) **A1**

(86) Date de dépôt PCT/PCT Filing Date: 2001/06/29

(87) Date publication PCT/PCT Publication Date: 2002/01/17

(85) Entrée phase nationale/National Entry: 2003/01/08

(86) N° demande PCT/PCT Application No.: DE 2001/002459

(87) N° publication PCT/PCT Publication No.: 2002/005368

(30) Priorité/Priority: 2000/07/12 (100 33 898.4) DE

(51) Cl.Int.⁷/Int.Cl.⁷ H01M 8/02, H01M 8/24, H01M 8/12

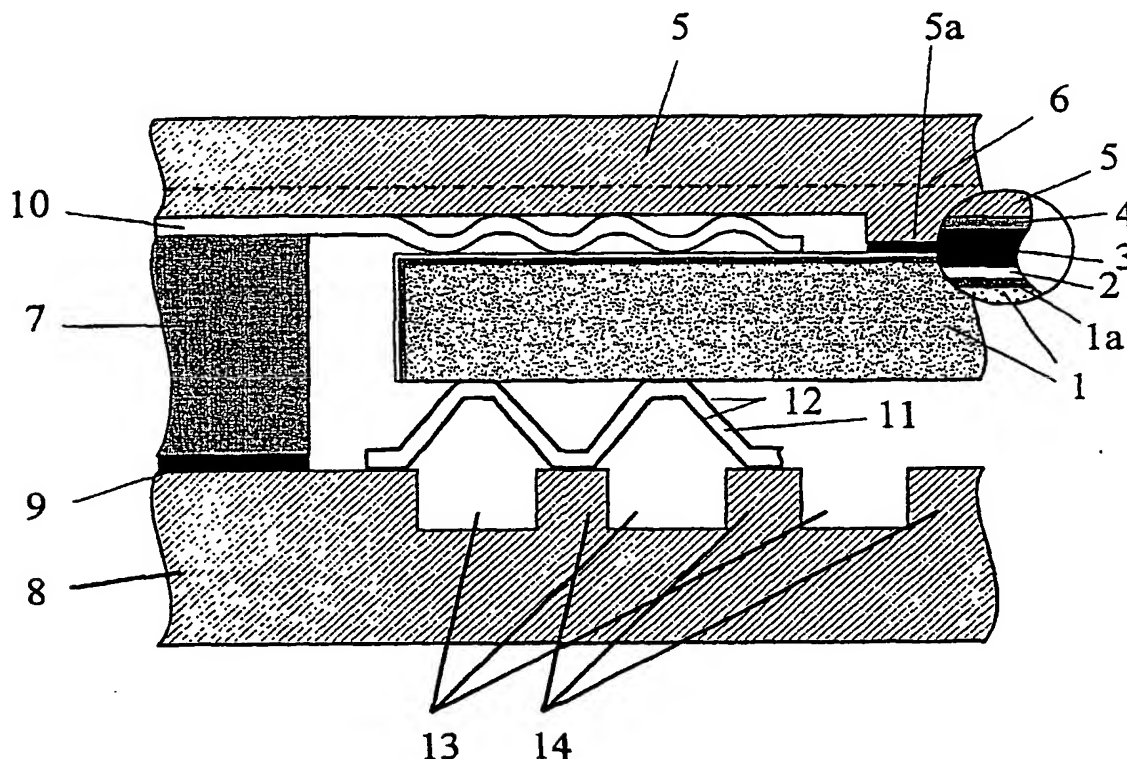
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(54) Titre : PILE A COMBUSTIBLE HAUTE TEMPERATURE

(54) Title: HIGH TEMPERATURE FUEL CELL



(57) Abrégé/Abstract:

The invention relates to a high temperature fuel cell, comprising an anode (1, 1a), an electrolyte (2), a cathode (3), a cathode interconnection (5) and an anode interconnection (8). At least one elastic mass (11) is arranged between the anode and anode interconnection to absorb relative movements.

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Abstract

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High Temperature Fuel Cell

The present invention relates to a high-temperature fuel cell.

A fuel cell incorporates a cathode, an electrolyte, and an anode. An oxidizing agent, e.g., air, is routed to the cathode, and fuel, e.g. hydrogen, is routed to the anode.

There are various types of fuel cells; these include the SOFC described in DE 44 30 958 C1, and the PEM fuel cell that is described in DE 195 31 852 C1.

The SOFC is also referred to as a high-temperature fuel cell since its operating temperature can reach 1000°C. Oxygen ions form on the cathode of a high-temperature fuel cell in the presence of the oxidizing agent. The oxygen ions diffuse through the electrolyte and recombine with the hydrogen that originates from the fuel to form water on the anode side. Electrons are liberated during this recombination, and electrical energy is generated thereby.

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As a rule, a plurality of fuel cells are connected together electrically and mechanically by connecting elements—also referred to as inter-connectors—in order to generate large amounts of electrical power. One example of a connecting element is the bipolar plate. Fuel cells that are stacked one above the other and electrically connected in series are formed using bipolar plates. This arrangement is referred to as a fuel-cell stack. The fuel-cell stacks comprise the interconnectors and the electrode-electrolyte units.

In addition to their electrical and mechanical characteristics, interconnectors incorporate gas-distribution structures. In the case of the bipolar plate, these are realized in the form of bars with electrode contact that separate the gas channels used to supply the electrodes from each other (DE 44 10 711 C1). Gas-distribution structures ensure that the operating medium is distributed evenly into the electrode spaces (the spaces in which the electrodes are located).

The following problems can arise with fuel cells and fuel-cell stacks:

- metal bipolar plates with a high aluminum content form Al_2O_3 covering layers that can act as an undesirable electrical insulator;

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- thermal stresses that are accompanied by relative movement of the individual components generally occur during cyclical temperature loads; these result from the various expansion behaviours or the different coefficients of expansion associated with the materials that are used during operation;
- in the prior art, glass bonds that are of low elasticity are used to seal the individual components of a fuel cell. Because of this, as a result of thermal stresses, there is a risk that cracks will form and there will be a loss of adhesion.

In this regard, in the prior art there is still no adequate compatibility between the comparatively high coefficients of expansion of, for example, the metallic bipolar plates and the electrode materials known at this time, the coefficients of expansion of which are comparatively small. On the one hand, thermal stresses can occur between electrodes and interconnectors, and these can result in damage within the fuel cells. On the other hand, this also applies to the glass bonds that are intended to ensure the integrity of the fuel cells, and that are frequently used in fuel cells.

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For this reason, it is the objective of the present invention to present a fuel cell in which long-term stable mechanical-electrical contact with the cathodes and anodes by the interconnectors is ensured. Problems that are based on thermal stresses, e.g., inadequate sealing, are to be precluded.

This objective has been achieved by a high-temperature fuel cell as defined in Claim 1. This comprises an anode, an electrolyte, and a cathode, as well as a cathode interconnector and an anode interconnector, there being at least one elastic medium arranged between the anode and the anode interconnector to absorb relative movements. Because of this, thermal stresses on the anode side that result from the different expansion behaviours of the individual components are balanced out.

It is advantageous that the cathode interconnector has a projecting surface that contacts the cathode (Claim 2). The effect of this projecting surface is that the cathode interconnector has an encircling edge, the height of which is smaller in cross section than it is on the cathode contact surface. The result of this is that there is a certain freedom of movement for the electrodes on the cathode side. The expansion behaviour of the materials, and in

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particular thermal stresses, can then be evened out better on the anode side and the cathode side.

It is particularly advantageous that there be an additional elastic medium between the cathode interconnector and the electrolyte, this being, in particular, a corrugated, perforated plate (Claim 3). Because of this, the electrodes and the electrolyte are freely suspended and particularly great freedom of movement is achieved for the electrodes. This means that the bending of the individual components that frequently occurs during operation is prevented. The perforated plate does not have to be corrugated overall, but can have a flat edge in order that it can be better stabilized by other components of the fuel cell.

The cathode can be of a smaller area than the anode (Claim 4). Because of this, on the surfaces that are not covered by the cathode the perforated plate can contact the electrolyte through its troughs so as to form a gas tight contact (Claim 5). This means that the cathode is protected against pressure and damage. Because of the gas-tight contact, it is possible to dispense with the glass ceramics that are used in the prior art in order to seal the gaps between the fuel cell and the interconnectors that separate the

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cathode space from the anode space. Because of their different expansion behaviours, these joints are particularly critical, and entail the above-discussed disadvantages with respect to the formation of cracks. The elasticity of the perforated plate can be varied with respect to its thickness, the angle of slope of the corrugations, and the number of corrugations. The profile of the corrugations is compressed between of the cathode interconnector and the electrolyte after assembly of the high-temperature fuel cell, so that ultimately pressure is exerted on the anode. This brings about the desired sealing effect between the cathode space and the anode space. The perforated plate can be of a high-temperature alloy, in particular an iron-chromium-aluminum alloy, e.g., Aluchrom[®] YHf (Material No. 1.4767), or a nickel based alloy, e.g., Nicrofer[®] 6025 HT (Material No. 2.4633). What is important is that the material be highly creep resistant and sufficiently elastic at high temperatures.

The cathode interconnector can be connected to the anode interconnector through a frame, so as to be electrically isolated from it (Claim 6). The frame fulfills the function of an insulating connecting element and a spacer for the interconnectors. When the high-temperature fuel cell is pressed together, the maximal force that can be exerted on the perforated plate on

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the cathode side and the additional elastic medium on the anode side is limited by the frame.

The frame can be connected to the perforated plate (Claim 7). To this end, the perforated plate should have a flat edge. The connection can be established by way of a soldering or welding process. Since, because of the projecting surface, the height of the cathode interconnector is smaller in cross section at its edge than at the cathode contact surface, on the edge in question, the cathode interconnector can contact the corrugated plate all around its flat edge. This results in mechanical stabilization of the perforated plate.

A glass ceramic layer can be arranged between the troughs of the perforated plate and the electrolyte (Claim 8). This layer serves to increase the tightness of the seal between the troughs and the electrolyte, and to create sealed and gas-tight electrode chambers.

It is preferred that the frame be of an iron-based alloy that contains aluminum (Claim 9). Such a frame can be annealed above 1000°C while air is being supplied to it. After this process, the surface of the frame has an

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electrically insulating aluminum oxide layer (Claim 10). The surface of the frame can thus very simply be made electrically insulating. A medium for insulating the frame can also be arranged between the frame and the anode interconnector (Claim 11). A glass ceramic layer can be used as such a medium (Claim 12). Since the frame and the interconnector exhibit very similar expansion behaviour at high temperatures, this location for a seal with a glass ceramic is less critical. However, it is also possible to use a layer of mica as a medium for insulating the frame electrically (Claim 13). This can be applied using a suitable paste technique. Because of its stratified structure, such a mica layer is sufficiently elastic, and this avoids the formation of cracks and a loss of adhesion during operation. If such an additional medium is used, one is not restricted to iron-based alloys that contain aluminum when selecting the material for the frame.

An electrically insulating medium can also be arranged between the frame and the anode (Claim 14). The electrically insulating medium is so arranged that it ensures electrical insulation between the frame and the anode and prevents short circuits. As a rule, the space between the frame and the anode is small once the high-temperature fuel cell has been assembled; this insulating medium may be necessary for this reason. The anode can have a

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glass-ceramic layer as the medium for electrical insulation (Claim 15). This can be applied to the sides that face the frame. Glass ceramics can be applied in a simple manner. However, insulation between the anode and the frame can also be ensured by the electrolyte. To this end, the anode must be coated with the electrolyte as far as the side that is opposite the cathode (Claim 16). Electrical insulation is ensured because of the material of the electrolyte that consists, for example, of yttrium-stabilized ZrO_2 .

According to Claim 1, at least one elastic medium for absorbing relative movement can be arranged between the anode interconnector and the anode. This can be a corrugated, elastic foil that incorporates openings (Claim 17). The material can consist of a high-temperature alloy, in particular an iron-chromium-aluminum alloy, e.g., Aluchrom[®] YHf, or a nickel-based alloy such as Nicrofer[®] 6025 HT. It is important that the material be highly creep resistant and sufficiently elastic at high temperatures. In addition, the foil incorporates openings. These openings can be punched out of the foil before it is shaped. The openings serve to supply the anode with fuel. If, for example, the anode is formed as a bipolar plate, the fuel flows out of gas channels, through the openings in the foil, and onto the anode.

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In one further embodiment of the present invention, provision is made such that the foil has nickel-aluminum alloys on both sides, at least in part (Claim 18). To this end, the elastic foil is joined on both sides to foils that contain 99% nickel. Because of the high temperature resistance of the nickel, the nickel-aluminum alloys ensure that the foil will possess good electrical conductivity, which will remain stable over a long period, on its surface.

It is advantageous that the anode interconnector contain aluminum (Claim 19). This results in additional possibilities for ensuring the electrical conductivity between the anode interconnector and the elastic medium or the anode. The anode interconnector can also contain nickel-aluminum alloys, at least in part (Claim 20). To this end, at least one foil that contains nickel must be connected to the contact surfaces of the anode interconnector for the anode by the formation of alloy (Claim 21). This can either be the shaped foil that has the nickel-aluminum alloys, the troughs of which are connected to the anode interconnector by the formation of alloy (Claim 22), or other nickel foils can be connected with the anode interconnector by the formation of alloy. The anode interconnector thus contains nickel aluminum alloys, at least in part, on the contact surfaces to the corrugated foil.

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The nickel-aluminum alloys can, for example, be nickel aluminides (e.g., NiAl, NiAl₂, Ni₃Al). Generally speaking, when used as contact layers in high-temperature fuel cells, such nickel-aluminum alloys offer the following advantages:

- Nickel-aluminum alloys act as diffusion barriers for alloy components of the steels used for interconnectors and other components of the fuel cell and avoid the formation of low-conductivity corrosion products (e.g., aluminum oxide) on boundary surfaces, for example, between the anode interconnector and the nickel coating of the foil.
- Nickel-aluminum alloys are resistant to high temperatures (e.g., the melting point of NiAl is 1638°C).
- Nickel-aluminum alloys are electrically conductive to a sufficient degree.
- Low material and machining costs.

The formation of insulating aluminum oxide layers is avoided because of the properties of the nickel-aluminum alloys. From this it follows that a reduction of contact resistance or a high level of conductivity on the anode

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side is achieved by the nickel aluminum alloys, and this leads to contact, particularly in the fuel-cell stack, that is stable over a long period.

It has been shown that hot-pressing methods at temperatures of up to 1150°C are particularly well-suited for producing nickel-aluminum alloys that are stable for a long period. Welding methods carried out in an atmosphere of protective gas as well as, to some extent, plasma spraying methods can also be used.

A further possibility for producing nickel-aluminum alloys is to use a galvanic nickel plating process. The nickel plated surface is subsequently annealed in a vacuum with the formation of nickel-aluminum alloys, preferably at 1150°C.

An elastic nickel grid can be arranged between the anode and the anode interconnector (Claimed 23). The nickel grid not only evens out relative movements between the anode and the interconnector; it also offers the added advantage that it ensures electrical contact with the anode evenly through the grid points of the grid itself and thus balances out the above-described disadvantageous manufacturing tolerances. It is also possible to

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have both elastic media, which is to say the nickel grid and the corrugated foil, arranged on the anode side.

Finally a cathode contact layer can be arranged between the cathode interconnector and the cathode (Claim 24). This can lie flush on the cathode. During the joining process, this layer can serve, on the one hand, to balance out tolerances, and on the other can serve as a diffusion barrier for chromium that is vapourizing out of the cathode interconnector.

A fuel cell stack comprises at least two such high-temperature fuel cells (Claim 25). This achieves higher power outputs.

The present invention will be described in greater detail below on the basis of two embodiments shown in the drawings appended hereto. These drawings show the following:

Figure 1: A cross-section through a high-temperature fuel cell with a corrugated foil that absorbs relative movements;

Figure 2: A cross-section through a high-temperature fuel cell with a nickel grid to absorb relative movements.

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On the right-hand side of both the drawings there is a circular cross-sectional enlargement that is provided to show more clearly those components that are located close together.

In Figure 1, the anode 1 consists of NiO and 8YSZ-stabilized ZrO₂. It is 1500 µm thick, so that the anode is configured as an anode substrate and performs a supporting function. The anode function layer 1a is 5 µm thick and is of the same material as the anode 1. The anode function layer 1a is of a lower porosity than the anode 1 so as to ensure an even coating with the electrolyte 2 of 8 YSZ. The anode 1 is completely coated with the electrolyte 2 down as far as the lower base surface. This coating is at least 5 µm thick. The electrolyte 2 is of a sufficiently low electrical conductivity to isolate the anode 1 from its adjacent components in the fuel cell as far as the anode interconnector 8. The area of the cathode 3 is smaller than the anode 1, and is arranged on the electrolyte 2. In its standard form, this consists of La_{0,66} Sr_{0,3} MnO₃ and its layer thickness is 40 µm; it consists, for example, of LaCoO₃ and it is 75 µm thick; it is shown only in the section

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enlargement. Amongst other things, manufacturing tolerances that originate during the production of bipolar plates or electrode-electrolyte units can be evened out by the cathode contact layer 4, so that low-conductivity contact points between the cathode 3 and the cathode interconnector 5 are avoided.

The cathode contact layer 4 is contacted through a projecting surface 5a of the cathode interconnector 5. Because of the projecting surface 5a, the cathode interconnector 5 has a circumferential edge, the height of which—in cross section—is smaller than on the aforementioned projecting surface 5a. A steel, Material No. 1.4742, can be used as the material for this. The cathode interconnector 5 is in the form of a bipolar plate. The gas channels are indicated by the dashed-dotted line 6. The flow of gas runs in the horizontal plane, for example, from left to right. On the edge in question, the cathode interconnector is connected to an anode interconnector through a frame 7 so as to be electrically insulating. The frame 7 can be of an iron-based alloy. It is connected through an electrically insulating layer 9 to the anode interconnector 8. On the cathode side, the frame 7 is connected to a perforated plate 10 which is flat on the edges and which possesses elastic properties and which, for the remainder, is corrugated; the troughs of this corrugated plate 10 contact the electrolyte 2 all round on the surfaces that are not covered by the cathode 3, so as to be gas tight. The crests of the

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perforated plate 10 contact the cathode interconnector 5 on its edge which, as discussed above, is of a lesser height in cross section than the projecting surface 5a, which contacts the cathode contact layer 4. The perforated plate 10 is of Nicrofer[®], and is 100 μm thick. However, the thickness can vary between 50 and 300 μm . All in all, the perforated plate 10 has four troughs to provide a gas-type contact, although more or fewer corrugations can be used. The gas-tight electrode chambers are formed thereby. On the contact surface with the anode interconnector 8, the frame 7 has an electrically insulating medium 9, e.g., of glass ceramic or mica. The anode interconnector 8 is also in the form of a bipolar plate, and incorporates gas channels 13 that are separated from each other by the bars 14. The operating media for the anode and cathode are thus supplied in a cross-flow pattern. A parallel gas feed (direct or counter-flow pattern) can also be used. A more homogenous temperature distribution across the fuel cell can be anticipated if a parallel gas feed is used. The bars 14 contact the anode. Between the anode interconnector 8 and the anode 1 there is an elastic medium 11 that is in the form of a corrugated elastic foil that incorporates openings. This absorbs relative movements between the anode and the anode interconnector, and serves to balance out the expansion of the individual components that is brought about by thermal stresses. The foil 11

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can be of Aluchrom[®] YHf or Nicrofer[®] 6025HT, and can be, for example, 100 μm thick, although this can vary between 50 and 300 μm . The openings serve to supply the anode 1 with fuel. The fuel flows out of the gas channels 13 in the anode interconnector 8, through the openings in the foil 11, to the anode 1. The foil contains nickel-aluminum alloys 12 on both sides to reduce contact resistance. The anode-side foil 11, the cathode-side perforated plate 10, and the projecting contact surface 5a of the cathode interconnector 5 thus ensure equalization of the v.a. thermal stresses that occur during recycling.

Figure 2 differs from Figure 1 with respect to the electrical insulation of the frame 17, the material of the frame itself, as well as the elastic medium 21 that is arranged between the anode interconnector 8 and the anode 1, so as to absorb relative movements.

In Figure 2, the frame 17 consists of an iron-based alloy that contains aluminum, e.g., Aluchrom[®] YHf. It is connected to the anode interconnector 8 and has an electrically insulating layer 19 of aluminum oxide on its upper surface. The proportion of 5% aluminum in the frame is sufficient to form this electrically insulating covering layer 19 of aluminum oxide by annealing

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at 1000°C during the addition of air. The insulating covering layer 19 completely covers the surface of the frame 17, and is for this reason shown as an enclosing layer. In Figure 2, frame 17 is sufficiently high that additional elastic medium for absorbing relative movements can be arranged between of the anode interconnector 8 and the anode 1. In Figure 2, this is an elastic nickel grid 21 that is 250 µm thick and has a mesh size of 200 µm. The diameter of the wire is 125 µm. The fuel flows out of the gas channels 13 of the anode interconnector 8, through the mesh of the nickel grid 21, to the anode 1.

The embodiments that are shown in Figure 1 and Figure 2 can be combined with each other without restriction. Thus, in Figure 1, there can be a frame 17 that contains aluminum, as is described with respect to Figure 2. On the other hand, in Figure 2, a frame 7 such as the one described in Figure 1 can be used .

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Patent Claims

1. High-temperature fuel cell, which comprises an anode (1), and electrolyte (2), and a cathode (3), as well as a cathode interconnector (5), characterized in that an elastic medium is arranged between the cathode interconnector (5) and the electrolyte (20).
2. High-temperature fuel cell as defined in Claim 1, characterized in that the cathode interconnector (5) has a projecting surface (5a) that contacts the cathode.
3. High-temperature fuel cell as defined in one of the preceding claims, characterized in that a further elastic medium (10), in particular a corrugated perforated plate, is arranged between the cathode interconnector (5) and the electrolyte (2).
4. High-temperature fuel cell as defined in one of the preceding claims, characterized in that the cathode (3) is of a smaller area than the anode (1).

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5. High-temperature fuel cell as defined in Claim 4, characterized in that the perforated plate (10) contacts the electrolyte (2) through its troughs on those surfaces not covered by the cathode (3), so as to be gas tight.
6. High-temperature fuel cell is defined in one of the preceding claims, characterized in that the cathode interconnector (5) is connected to the anode interconnector (8) through a frame (7, 17) so as to be electrically insulated therefrom.
7. High-temperature fuel cell as defined in Claim 6, characterized in that the frame (7, 17) is connected to the perforated plate (10).
8. High-temperature fuel cell as defined in one of the Claims 3 to 7, characterized in that a glass ceramic layer is arranged between the troughs of the perforated plate (10) and the electrolyte (2).
9. High-temperature fuel cell as defined in one of the claims 6 to 8, characterized in that the frame (7, 17) is of an iron-based alloy that contains aluminum.

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10. High-temperature fuel cell as defined in Claim 9, characterized in that the surface of the frame (7, 17) has an electrically insulating layer of aluminum oxide (19).
11. High-temperature fuel cell as defined in one of the claims 6 to 10, characterized in that a medium (9) for insulating the frame (7, 17) electrically is arranged between the frame (7, 17) and the anode interconnector (8).
12. High-temperature fuel cell as defined in Claim 11, characterized in that a glass ceramic layer is used as the medium (9).
13. High-temperature fuel cell as defined in Claim 11, characterized in that a layer of mica is arranged as the medium (9).
14. High-temperature fuel cell as defined in one of the claims 6 to 13, characterized in that a medium for electrical insulation is arranged between the frame (7, 17) and the anode (1).

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15. High-temperature fuel cell as defined in Claim 14, characterized in that the anode has a glass ceramic layer as the electrically insulating medium.
16. High-temperature fuel cell as defined in Claim 14, characterized in that the anode (1) is coated with the electrolyte (2) as far as the side that is opposite the cathode.
17. High-temperature fuel cell as defined in one of the preceding claims, characterized in that a corrugated elastic foil (11) that incorporates openings is arranged between the anode (1) and the anode interconnector (8).
18. High-temperature fuel cell as defined in Claim 17, characterized in that the foil (11) incorporates nickel-aluminum alloys, at least in part, on both sides.
19. High-temperature fuel cell is defined in one of the preceding claims, characterized in that the anode interconnector (8) contains aluminum.

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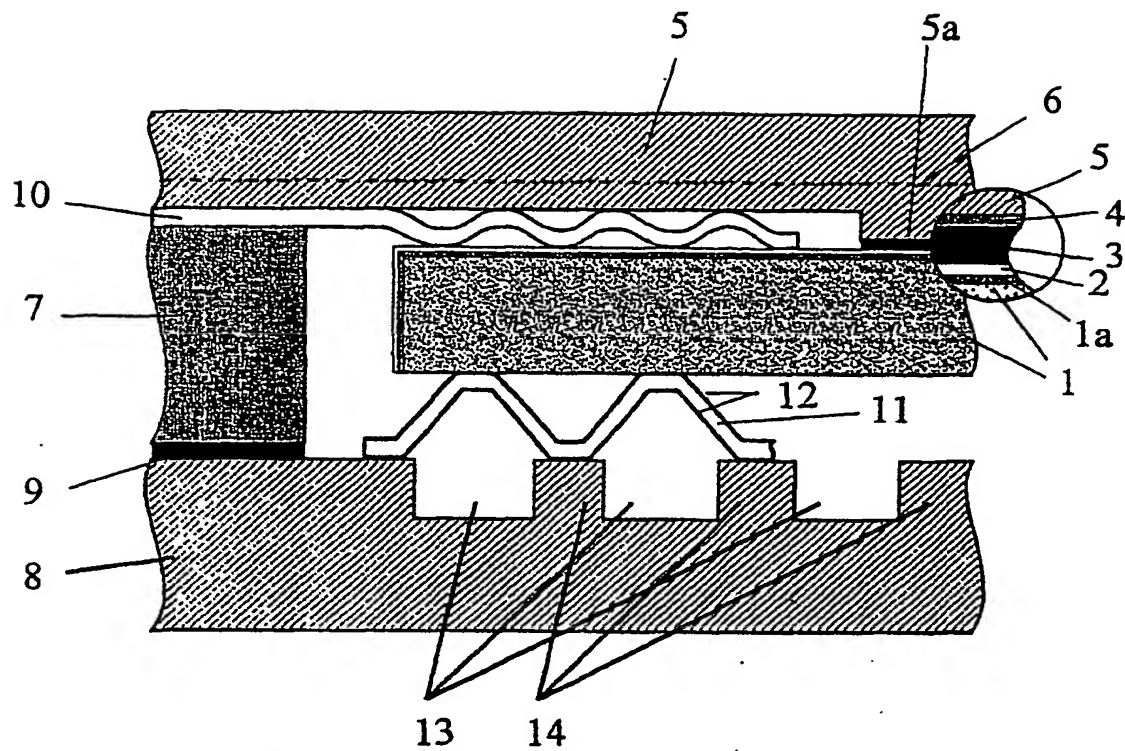
20. High-temperature fuel cell as defined in one of the preceding claims, characterized in that the anode interconnector (8) has nickel-aluminum alloys, at least in part.
21. High-temperature fuel cell as defined in one of the preceding claims, characterized in that at least one foil that contains nickel is connected to the contact surfaces of the anode interconnector (8) for the anode by the formation of alloy.
22. High-temperature fuel cell as defined in one of the Claims 17 to 21, characterized in that the troughs of the corrugated foil (11) are connected with the anode interconnector (8) by the formation of an alloy.
23. High-temperature fuel cell as defined in one of the preceding claims, characterized in that an elastic nickel grid (21) is arranged between the anode (1) and the anode interconnector (8).
24. High-temperature fuel cell as defined in one of the preceding claims, characterized in that a cathode contact layer (4) is arranged between the anode interconnector (5) and the cathode (3).

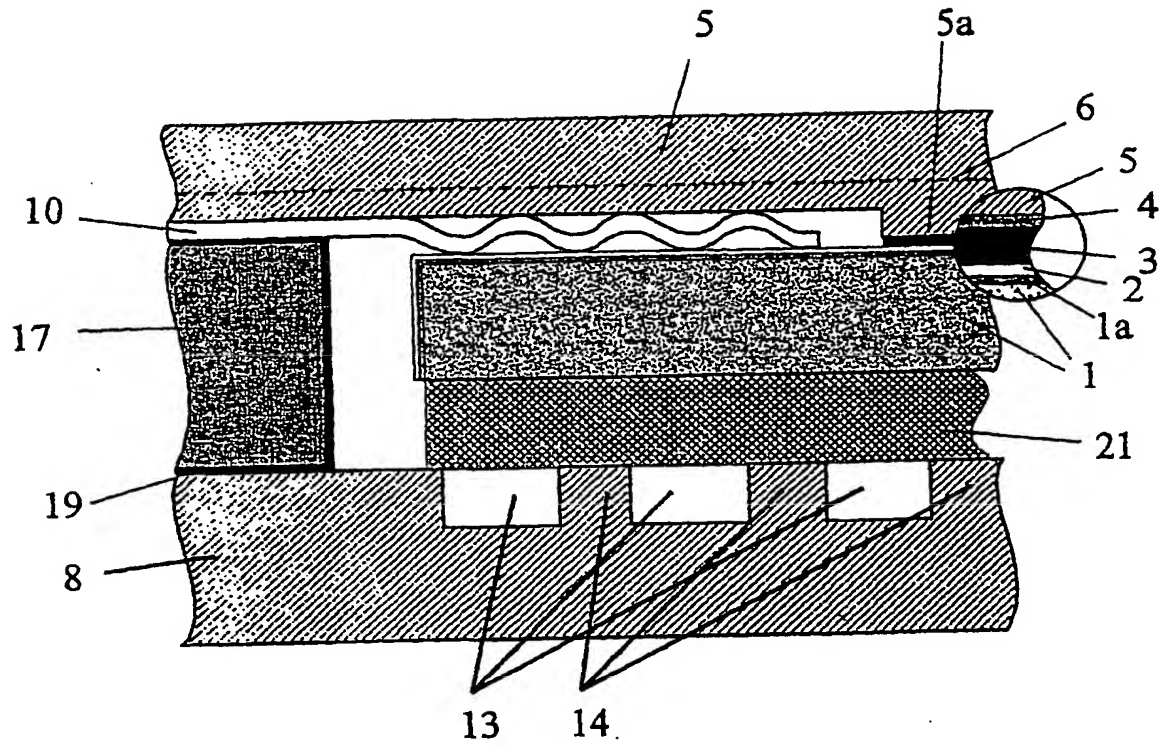
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25. Fuel cell stack, comprising at least two high-temperature fuel cells as defined in one of the preceding claims.

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**Fig. 1**

**Fig. 2**

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